Determination of Organophosphorus Pesticide Residues in Greek Virgin Olive Oil by Capillary Gas Chromatography

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In this study, the occurrence of 15 organophosphorus pesticide residues in Greek virgin olive oil was investigated. Analysis was carried out using capillary gas chromatography with specific detectors (FPD and NPD), after sample extraction with *n*-hexane and cleanup by partitioning between *n*-hexane and acetonitrile. Sixty-two samples of virgin olive oil were taken from the major production areas and packing companies of Greece during 1992−1994. In 46 samples, 9 organophosphorus pesticides, namely dimethoate, fenthion, omethoate, chlorpyrifos, methamidophos, parathion-methyl, parathion, methidathion, and malathion, were found, in concentrations ranging from 0.0005 to 0.1800 mg/kg. Diazinon, pirimiphos-methyl, paraoxon-methyl, malaoxon, carbophenothion, and azinphos-ethyl were not detected in any sample. In most samples pesticide residues were below the detection limits (0.0001 and 0.001 mg/kg), and most of the positive findings were a fraction (i.e., <0.09−18%) of the FAO/WHO Codex Alimentarius maximum residue limits (MRLs) except for dimethoate, which was ranged between 1 and 45%. Only one sample contained dimethoate residue that exceeded the Codex MRL for refined olive oil.

Keywords: *Virgin olive oil; pesticide residues; organophosphorus insecticides; capillary gas chromatography*

INTRODUCTION

Olive oil is obtained from the fruit of the olive tree (*Olea europea*) that grows mainly in the Mediterranean countries. Virgin olive oil is the oil obtained from the fruit of the olive tree by mechanical or other physical processes, mainly under thermal conditions which do not alter the olive oil quality (EC, 1991).

Because of its nutritional and biological characteristics, it is one of the most important components of the Mediterranean diet (Ferro-Luzzi and Sette, 1989). Greece is among the leading oil olive producers internationally, and the mean consumption of olive oil in Greece is of 20 kg per person per year (Kiritsakis, 1988).

Olive trees are attacked by several pests, mainly the olive fruit fly Bactocera (Dacus) Oleae, and receive treatment with several pesticides. Those more extensively used belong to the class of organophosphorus insecticides and are mainly fenthion, dimethoate, diazinon, parathion-methyl, methidathion, and azinphos-ethyl. From existing data 100−120 tonnes of fenthion and 80−90 tonnes of dimethoate are used in Greece annually (Lentza-Rizos and Avramides, 1991).

The presence of toxic residues in olive oil has been reported by several researchers (Lentza-Rizos and Avramides, 1994; Morchio et al., 1992). Because pesticide residues in food constitute a significant health risk and olive oil has a high consumption rate among people of the producing countries, the continuous control of pesticide residues in olive oil is of great importance.

The Codex Alimentarius Commission of the Food and Agriculture Organization of the United Nations (FAO) and the World Health Organization (WHO) has established maximum residue limits (MRLs) for pesticide residues in olives and olive oil (Table 1) (Codex Alimentarius Commission, 1996). Maximum levels for pesticide residues in and on certain products of plant origin, including olives, are also fixed by the European Community (EC, 1976).

Many multiresidue procedures employing different cleanup techniques and a variety of detection methods have been reported for the determination of organophosphorus pesticide residues in olive oil. In brief, they can be classified into procedures using gas chromatography (GC) after a cleanup step based on partitioning between hexane or light petroleum and acetonitrile (AOAC, 1984; FDA, 1982), gel permeation chromatography (Specht, 1987), and solid-phase extraction using a multicartrige system (Di Muccio et al., 1990; Gillespie et al., 1995). The main disadvantage of these methods is the small sample manipulation (1−3 g) resulting in low analyte detectability.

Direct injection into the GC, using a glass precolumn with an oil recovery tank, was found to be effective for separating organophosphorus pesticides from vegetable oils (Morchio et al., 1992). However, the need for limited commercially available equipment makes this technique impractical.

More recently, an on-line combination of gel permeation chromatography and GC has been developed to determine organophosphorus pesticides in olive oil (Vreuls et al., 1996). Although this method is promising, it appeared that it is not yet robust enough for routine analysis.

An improvement of the hexane−acetonitrile partitioning step of the AOAC official method (AOAC, 1984) has been reported for the determination of fenthion and its metabolites, allowing the use of larger sample

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Table 1. Codex Alimentarius MRLs for Olives and Olive Oil

	MRL (mg/kg)				
	olives			oil	
pesticide	fresh	processed	not specified	virgin	refined
carbaryl	10	1			
carbophenothion				0.2[a]	
deltamethrin			0.1		
dimethoate	1	0.05			0.05
fenthion and metabolites			1	1	
methidathion			2	2	
paraquat			1		
parathion			0.5	2	
permethrin (sum of isomers)			1		
pirimiphos-methyl			5		

[a] Withdrawn by the Codex Alimentarius Commission in 1994.

quantities (Lentza-Rizos and Avramides, 1990). This procedure has also been performed for the insecticides diazinon, dimethoate, methidathion, parathion-methyl, parathion, chlorpyrifos, and azinphos-ethyl using GC with nitrogen−phosphorus detection (NPD) and packed columns (Lentza-Rizos, 1994). Although this procedure is quite simple, the low separation power of packed columns and the remarkable changes with time of NPD sensitivity, due to degradation of the bead in the detector (Vreuls et al., 1996), along with its limited selectivity (Lawrence, 1987), complicate the trace level determination of organophosphorus compounds.

In this paper, the further improvement of the above method, by the combination of the high separation power of capillary GC columns and the high sensitivity and selectivity of flame photometric detection (FPD), for the determination of 15 organophosphorus pesticides with different polarities and water solubilities, wider than those previously studied, in olive oil, is reported.

The target compounds studied, namely methamidophos, omethoate, diazinon, dimethoate, pirimiphos-methyl, paraoxon-methyl, chlorpyrifos, malaoxon, parathion-methyl, malathion, fenthion, parathion, methidathion, carbophenothion and azinphos-ethyl, were selected for monitoring in Greek virgin olive oil, due to their intensive use in olive tree treatment. They are also included in the pesticides for which MRLs have been established by the European Communities for olives and by the Codex Alimentarius Commission for olive oil.

Sixty-two samples of Greek virgin olive oil were analyzed according to the proposed optimized method. Determination was carried out by capillary GC using FPD in the phosphorus specific mode. Confirmation was applied in the samples of positive findings using the specific NPD with capillary column of different polarity. The results of this monitoring of virgin olive oil samples from Greece are reported.

MATERIALS AND METHODS

Sampling. Forty-eight samples of virgin olive oil were collected from some of the major olive oil production areas of Greece, namely Lasithi (Crete), Hania (Crete), Messinia (Peloponissos), and Rodos, during 1992−1993 (26 samples) and 1993−1994 (22 samples). Virgin olive oil samples were collected in dark glass bottles. The samples were transported to the laboratory and kept for a short time at 4 °C until analysis. Fourteen samples of commercially packed Greek virgin olive oil were also analyzed during the same time period.

Reagents. *n*-Hexane and acetone were products of Lab scan (pestiscan) Dublin, (Ireland). Acetonitrile was of HPLC grade, a product of Carlo Erba, Milano (Italy). Water was a product of Reidel-de Haen (pestanal), Seelze (Germany). Methamidophos, dimethoate, pirimiphos-methyl, malathion, methidathion, and carbophenothion of purity 97−99% were also products of Riedel de Haen (pestanal). Diazinon of purity 99.4% was a product of Ciba-Geigy. Parathion-methyl, chlorpyrifos, omethoate, paraoxon-methyl, malaoxon, fenthion, parathion, and azinphos-methyl of purity 95−99.8% were products of Dr. Ehrenstorfer, Augsburg (Germany). Stock solutions of each organophosphorus pesticide were prepared in acetone at 1000 μg/mL. A solution of the mixture of all pesticides was also made up in acetone containing 0.1−1 μg/mL from each one.

Extraction and Cleanup. The extraction method employed was based on the procedure of Lenza-Rizos and Avramides (1990). A 10-g sample of oil was mixed with 50 mL of *n*-hexane saturated with acetonitrile. The mixture with 100 mL of acetonitrile saturated with *n*-hexane and 1 mL of water was shaken gently for 1 min and left for 15 min to allow the phases to separate. The lower acetonitrile phase was run into a second separating funnel containing 25 mL of *n*-hexane saturated with acetonitrile and the above procedure for phase separation repeated. A second 50-mL volume of acetonitrile saturated with *n*-hexane was added to the first separating funnel, followed by 0.5 mL of water, and the extraction procedure was repeated. The compined acetonitrile phases were rotary evaporated (bath temperature, 30 °C) to ≈1 mL, 5 mL of acetone was added, and the evaporation was repeated to almost dryness. The residue was redissolved in acetone and was transferred quantitatively into a 2-mL volumetric flask for the GC analysis.

GC Analysis. The analysis of the 15 organophosphorus pesticides was carried out by capillary GC using the following instruments: (1) Carlo Erba model Mega 2 gas chromatograph with FPD (model 50), equipped with a 526-nm interference filter (phosphorus mode), split/splitless injection port, a DB-1701 fused silica capillary column by J&W Scientific Inc. (30 m × 0.53 mm i.d., 1 μm film thickness) and autosampler model A200S, with program for the evaluation of GC runs (Chrom-Card, Fisons Instruments, Rodano, Italy); (2) Hewlett-Packard model 5890 II GC, with NPD, split/splitless injection port, a CP-SIL 13CB fused silica capillary column by Chrompack (50 m × 0.32 mm i.d., 0.4 μm film thickness) and autosampler model 7673, with program for the evaluation of GC runs (HPCHEM, Hewlett-Packard Co., Palo Alto, CA).

The temperature program applied in GC/FPD was as follows: 100 °C for 1 min, 100−140 °C at 35 °C/min, 140−220 °C at 5 °C/min, 220−260 °C at 10 °C/min, and 260 °C for 30 min. The temperature of the detector was 250 °C. The injection was carried out splitless at 250 °C, and the injection volume was 1 μL.

The temperature program applied in GC/NPD was as follows: 100 °C for 1 min, 100−150 °C at 30 °C/min, 150 °C for 2 min, 150−205 °C at 3 °C/min, 205−260 °C at 2 °C/min, and 260 °C for 1 min. The temperature of the detector was 280 °C. The injection was carried out splitless at 250 °C, and the injection volume was 1 μL.

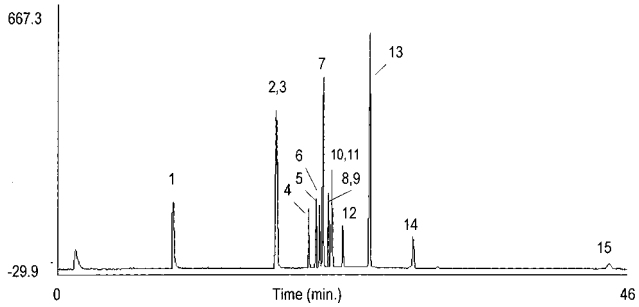

Figure 1. Gas chromatogram of 15 organophosphorus pesticides on a DB-1701 column with FPD: (1) methamidophos; (2) omethoate; (3) diazinon; (4) dimethoate, (5) pirimiphos-methyl; (6) paraoxon-ethyl; (7) chlorpyrifos; (8) malaoxon; (9) parathion-methyl; (10) malathion; (11) fenthion; (12) parathion; (13) methidathion; (14) carbophenothion; (15) azinphos-ethyl.

Table 2. RRTs of 15 Organophosphorus Pesticides on Two Columns Detected with FPD and NPD Detectors

organophosphorus pesticide	RRT	
	DB-1701/FPD	CP-SIL 13CB/NPD
methamidophos	0.393	0.195
omethoate	0.753	0.510
diazinon	0.753	0.719
dimethoate	0.876	0.677
pirimiphos-methyl	0.904	0.938
paraoxon-ethyl	0.917	0.776
chlorpyrifos	0.928	0.987
malaoxon	0.949	0.887
parathion-methyl	0.949	0.874
malathion	0.961	0.979
fenthion	0.961	1.005
parathion	1.000	1.000
methidathion	1.086	1.196
carbophenothion	1.230	1.508
azinphos-ethyl	1.888	2.076

RESULTS AND DISCUSSION

Determination. The target pesticides were determined by GC using capillary columns and specific detectors; FPD (P mode) and NPD. Their relative retention times (RRTs), with respect to parathion, on the DB-1701 and CP-SIL 13CB columns, respectively, are given in Table 2.

The GC/FPD and GC/NPD chromatograms of a standard mixture containing azinphos-ethyl at 0.1 μg/mL; dimethoate, parathion-methyl, fenthion, and parathion at 0.2 μg/mL; malathion and carbophenothion at 0.3 μg/mL; methamidophos, omethoate, and diazinon at 0.4 μg/mL; pirimiphos, paraoxon-methyl, and malaoxon at 0.5 μg/mL; and chlorpyrifos and methidathion at 1 μg/mL are shown in Figures 1 and 2, respectively. As can be seen from Figure 1, omethoate (**2**) and diazinon (**3**), malaoxon (**8**) and parathion-methyl (**9**), and malathion (**10**) and fenthion (**11**) coelute from the DB-1701 column. On CP-SIL 13CB the peaks corresponding to all analytes are satisfactorily resolved (Figure 2).

The matrix interference during analysis of olive oil samples in the GC/FPD system was very limited in comparison with that in the GC/NPD system. Gas chromatograms of spiked olive oil samples in GC/FPD were indistinguishable from those obtained with the standard solution of pure pesticides. Figure 3 compares the two detectors for extracts of olive oil, containing 0.002 mg/kg fenthion. The gas chromatogram of the FPD shows better baseline stability and fewer peaks than the nitrogen−phosphorus one, although both detectors easily made the determination of the indicated organophosphate possible.

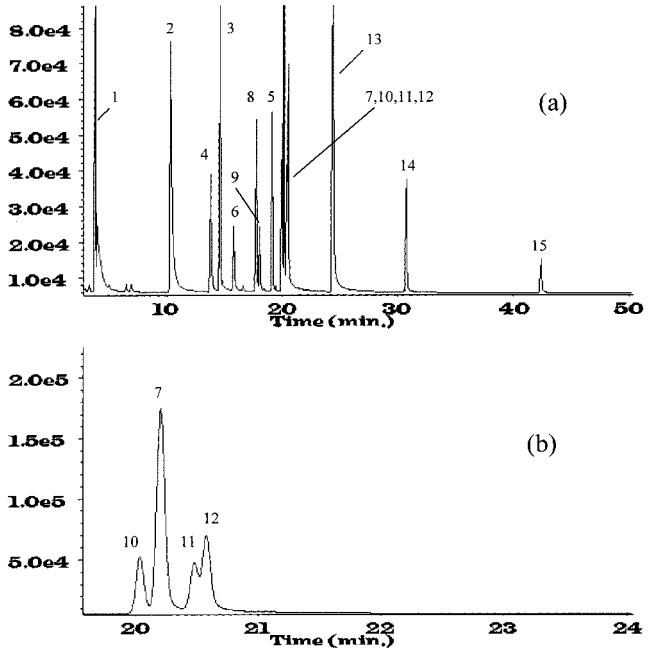

Figure 2. (a) Gas chromatogram of 15 organophosphorus pesticides on a CP-SIL 13CB column with NPD: (1) methamidophos; (2) omethoate; (3) diazinon; (4) dimethoate; (5) pirimiphos-methyl; (6) paraoxon-ethyl; (7) chlorpyrifos; (8) malaoxon; (9) parathion-methyl; (10) malathion; (11) fenthion; (12) parathion; (13) methidathion; (14) carbophenothion; (15) azinphos-ethyl. (b) Magnification of a part of (a), showing satisfactory resolution for pesticides **7** and **10**−**12**.

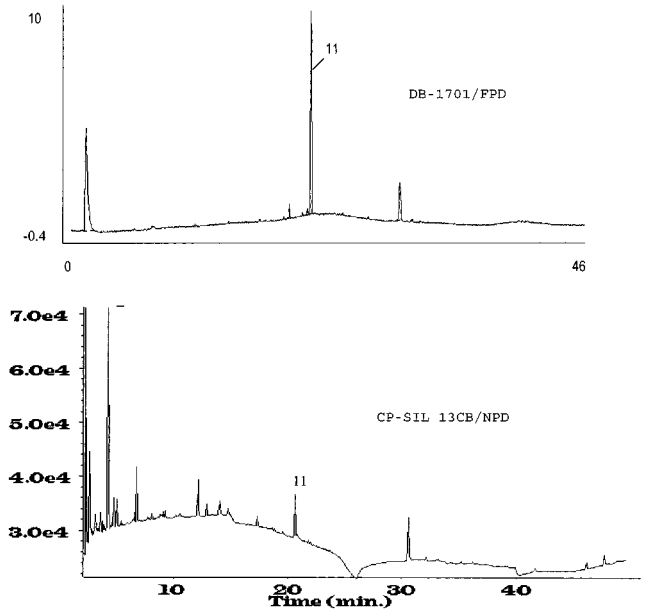

Figure 3. Gas chromatograms of an extract of virgin olive oil containing fenthion with FPD and NPD using a DB-1701 and a CP-SIL 13CB column, respectively.

The calculation of the amount of the organophosphorus pesticides present was carried out using the FPD in phosphorus mode by comparing the peak areas for unknown samples with the corresponding peaks for standards, according to established procedures. As the matrix interference during analysis of olive oil samples in the GC/FPD system was insignificant, the preparation of calibration standard solutions in control sample extracts was not necessary. The results were confirmed with the NPD on a CP-SIL 13CB column. In case one or more of the pesticide pairs that were not resolved on

Table 3. Mean Percent Recovery ± RSD of 15 Organophosphorus Pesticides in Olive Oil Samples at 0.01, 0.1, and 1 mg/kg Fortification Levels (n = 3)

	mean % recovery ± RSD at fortification level of			
organophosphorus pesticide	0.01 mg/kg FPD	0.1 mg/kg FPD	0.1 mg/kg NPD	1 mg/kg FPD
methamidophos	82 ± 5	81 ± 7	134± 22	89 ± 4
omethoate	80 ± 5	81 ± 5	166 ± 25	93 ± 5
diazinon	81 ± 9	105 ± 8	82 ± 15	82 ± 6
dimethoate	97 ± 6	98 ± 5	155 ± 20	89 ± 7
pirimiphos-methyl	79 ± 5	81 ± 6	83 ± 11	87 ± 6
paraoxon-ethyl	81 ± 5	90 ± 6	97 ± 10	87 ± 3
chlorpyrifos	95 ± 7	88 ± 5	93 ± 12	90 ± 6
malaoxon	114 ± 4	87 ± 4	100 ± 14	87 ± 6
parathion-methyl	101 ± 2	82 ± 3	87 ± 10	84 ± 3
malathion	108 ± 5	83 ± 8	85 ± 12	86 ± 5
fenthion	82 ± 2	94 ± 2	95 ± 12	89 ± 3
parathion	85 ± 4	82 ± 2	84 ± 15	96 ± 5
methidathion	79 ± 5	96 ± 3	121 ± 18	84 ± 4
carbophenothion	103 ± 8	84 ± 2	73 ± 11	92 ± 4
azinphos-ethyl	78 ± 3	74 ± 5	118 ± 13	73 ± 5

the DB-1701 column were present, identification was performed with the CP-SIL 13CB column. Quantification was carried out by NPD only when both components of each pair were identified; otherwise, it was based on FPD.

Recovery Experiments and Detection Limits (DLs) of Target Analytes. Recovery experiments, concerning the 15 organophosphorus pesticides, were performed in olive oil samples, at various fortification levels. Data derived from these experiments (in triplicate) are presented in Table 3. The mean recoveries, at three fortification levels (0.01, 0.1, and 1 mg/kg), obtained by FPD, ranged from 73% to 114% for all of the analytes, approaching complete recovery in most cases. As can be seen, the recovery values were not related to the spiking level. The mean recoveries of olive oil samples fortified at the 0.1 mg/kg level, obtained by NPD, are presented in Table 3. The majority of analytes were recovered with values ranging between 73% and 121%. The high recovery values of methamidophos, omethoate, and dimethoate (134%, 166%, and 155%, respectively) could be attributed to matrix effects. Similar observations were reported by others (Lentza-Rizos and Avramides, 1994). The precision of the GC/FPD method expressed by the relative standard deviation (RSD) of the mean recovery values, when triplicate spiked olive oil samples were analyzed, was better than 9%. The RSD values for all compounds in the GC/NPD system ranged from 10% to 25%. The results from this comparison study suggest that FPD is better suited for the detection of organophosphorus pesticides than NPD.

Table 4 shows the DLs of the target compounds determined after olive oil samples were spiked at lower concentration levels. The DLs, calculated by using a signal-to-noise (S/N) ratio of 3, were in the range of 0.0001−0.001 mg/kg, far below the MRLs for pesticide residues in olive oil established by FAO/WHO (Table 1). These concentrations correspond to amounts of 0.5−5 pg injected into the GC/FPD system. This is surprisingly close to the detection limits of 1−1.5 pg reported by others (Morchio et al., 1992) using direct injection of 1:1 diluted (with acetone) olive oil sample into the GC system with the same type of FPD as in the present work (Carlo-Erba FPD, model 50).

Monitoring Data. A study of the possible contamination of virgin olive oil with the 15 target compounds was carried out during the crop years 1992−1993 and 1993−1994. Detectable levels of organophosphorus pesticides were found.

A. Samples from Individual Growers. Forty-eight samples of virgin olive oil, collected from individual growers at the locations mentioned, were analyzed. The levels of the 15 organophosphorus pesticides in the virgin olive oil samples are presented in Table 5.

Thirteen samples contained no detectable residues. Dimethoate was detected in 29 samples at concentrations ranging from 0.0005 to 0.0816 mg/kg. Fenthion residues were detected in 17 samples at concentrations ranging from 0.0009 to 0.1802 mg/kg. Omethoate was detected in 5 samples at concentrations ranging from 0.0007 to 0.0072 mg/kg. Chlorpyrifos was detected in 4 samples at concentrations ranging from 0.0072 to 0.1240 mg/kg. Methamidophos, parathion-methyl, and parathion were detected in 3 samples each. Methidathion and malathion were detected in 2 and 1 samples, respectively. Diazinon, pirimiphos-methyl, paraoxon-methyl, malaoxon, carbophenothion, and azinphos-ethyl were not detected in any sample.

B. Commercially Packed Oil. Fourteen samples of commercially packed virgin olive oil were analyzed. The levels of the 15 organophosphorus pesticides in the virgin olive oil samples are presented in Table 6.

Three samples contained no detectable residues. Dimethoate was detected in 6 samples at concentrations ranging from 0.0040 to 0.0175 mg/kg. Fenthion resi-

Table 4. DLs for 15 Organophosphorus Pesticides in GC/FPD

no.	compound	DL, mg/kg
1	methamidophos	0.0001
2	omethoate	0.0001
3	diazinon	0.0005
4	dimethoate	0.0001
5	pirimiphos-methyl	0.0001
6	paraoxon-ethyl	0.0005
7	chlorpyrifos	0.0001
8	malaoxon	0.001
9	parathion-methyl	0.0001
10	malathion	0.0005
11	fenthion	0.0001
12	parathion	0.0001
13	methidathion	0.0005
14	carbophenothion	0.0005
15	azinphos-ethyl	0.001

Table 5. Organophosphorus Pesticides in 48 Samples of Greek Virgin Olive Oil from Individual Growers, Produced during 1992/1994[a]

pesticide	mean value (mg/kg)	concn range (mg/kg)	no. of positive samples
methamidophos	0.0011	0.0005−0.0015	3
omethoate	0.0023	0.0007−0.0072	5
diazinon		ND[b]	
dimethoate	0.0065	0.0005−0.0816	29
pirimiphos-methyl		ND	
paraoxon-methyl		ND	
chlorpyrifos	0.0427	0.0072−0.1240	4
malaoxon		ND	
parathion-methyl	0.0335	0.0039−0.0903	3
malathion	0.0197	0.0197	1
fenthion	0.0473	0.0009−0.1802	17
parathion	0.0696	0.0021−0.1558	3
methidathion	0.0063	0.0026−0.0100	2
carbophenothion		ND	
azinphos-ethyl		ND	

[a] Number of samples without residues: 13. Number of samples with residues greater than the Codex MRL: 1. [b] ND = not detectable.

Table 6. Organophosphorus Pesticides in 14 Samples of Commercially Packed Greek Virgin Olive Oil[a]

pesticide	mean value (mg/kg)	concn range	no. of positive samples
methamidophos		ND[b]	
omethoate		ND	
diazinon		ND	
dimethoate	0.0102	0.0040−0.0175	6
pirimiphos-methyl		ND	
paraoxon-ethyl		ND	
chlorpyrifos		ND	
malaoxon		ND	
parathion-methyl	0.0055	0.0020−0.0090	2
malathion		ND	
fenthion	0.0443	0.0030−0.1430	11
parathion	0.0070	0.0020−0.0170	3
methidathion	0.0030	0.0330	1
carbophenothion		ND	
azinphos-ethyl		ND	

[a] Number of samples without residues: 3. Number of samples with residues greater than the Codex MRL: 0. [b] ND = not detectable.

dues were detected in 11 samples at concentrations ranging from 0.0030 to 0.1430 mg/kg. Parathion, parathion-methyl, and methidathion were detected in 3, 2, and 1 samples, respectively. Methamidophos, omethoate, diazinon, pirimiphos-methyl, paraoxon-methyl, chlorpyriphos, malaoxon, malathion, carbophenothion, and azinphos-ethyl were not detected in any sample.

CONCLUSIONS

Because the treatment history of olive oil samples is, in most cases, unknown, our monitoring program was designed to check for as many pesticides as possible. For that reason a routine multiresidue method with low DLs has been improved and evaluated. This method is effective for the determination of the 15 most important organophosphorus pesticides, of different polarities and water solubilities, in olive oil, and at the same time it is quick and of low cost. The use of capillary GC with FPD in monitoring unknown olive oil samples for a large number of organophosphorus pesticides is preferred over NPD, because of its high selectivity and sensitivity.

The results of our monitoring on organophosphorus pesticides indicate that among 62 samples of virgin olive oil that were examined only 1 sample was found to have dimethoate (0.0816 mg/kg) in concentration above the MRL of Codex Alimentarius for refined olive oil (0.05 mg/kg). None of the commercially packed virgin olive oil samples contained residues higher than Codex MRLs.

The most common organophosphorus pesticide residues found in this study were fenthion and dimethoate. Fenthion was detected in 35% of olive oil samples from individual growers, ranging from 0.09% to 18% of the Codex MRL, and in 78% of commercially packed olive oil samples ranging from 0.3% to 14.3% of the MRL. The concentration range for fenthion residues, concerning the commercially packed olive oil samples, was similar to that observed by Lentza-Rizos during 1991−1992 (Lentza-Rizos and Avramides, 1994). Dimethoate was detected in 60% of the samples from individual growers, ranging from 1% to 45% of the Codex MRL for refined olive oil, and in 43% of commercially packed olive oil samples, ranging from 8% to 35% of the MRL. These findings, which were not presented in previous monitoring studies, indicate that dimethoate is one of the most important residues in olive olive.

However, the concentration levels found are below the Codex Alimentarious MRLs for all pesticides of the Codex list concerning olive olive (Table 1).

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